Superparamagnetic Behavior in Fe Ultrathin Films on GaN(0001)

P. K. J. Wong[1,3], W. Zhang[1], X. G. Cui[2], I. G. Will[1], Y. B. Xu[1,*], Z. K. Tao[2], X. Li[2], Z. L. Xie[2] and R. Zhang[2]
[1]Spintronics and Nanodevice Laboratory, Department of Electronics, University of York, York, YO10 5DD, United Kingdom
[2]Jiangsu Provincial Key Laboratory of Advanced Photonic and Electronic Materials and Department of Physics, Nanjing University, Nanjing 210093, People's Republic of China
[3]MESA$^+$ Institute for Nanotechnology, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands
*Correspondence author: yx2@ohm.york.ac.uk

Here we report on the studies on the growth, structural and magnetic properties of ultrathin Fe grown on GaN(0001) by molecular beam epitaxy. The films and their surfaces were monitored by *in-situ* reflection high energy electron diffraction (RHEED) and a crystal thickness monitor. The magnetic properties of the samples were determined by a superconducting quantum interference device (SQUID) magnetometer. The 2 μm thick GaN films used were prepared by metal-organic chemical vapor deposition (MOCVD) with a 20 nm low temperature (LT) buffer layer on c-Al_2O_3(0001). Prior loading into vacuum, the GaN surface was cleaned in a 1:1 HCl:H_2O solution then a 1:99 HF:H_2O solution for 2 min each with DI water rinses between and after etching. This *ex-situ* chemical processes combined with a vacuum anneal are useful to remove oxygen and carbon contaminants on the GaN surface [1]. After annealing at 600 ˚C for 1 h followed by cooling to room temperature (RT), streaky and sharp bulk-terminated (1×1) RHEED patterns were obtained, indicating a smooth GaN surface. Fe films with various thicknesses ranging from 2.5 to 150 ML were grown at RT at a chamber pressure of 1×10^{-10} mbar by *e*-beam evaporation at a rate of 2 Åmin^{-1}. Before samples retrieval for *ex-situ* characterization, the samples were capped with a 3.0 nm thick Au layer.

Evolution of the Fe(110) growth has been studied *in-situ* with the RHEED patterns as a function of Fe film thickness with the incident *e*-beam along the $[11\bar{2}0]$ of the GaN(0001). The streaky (1×1) pattern of the clean GaN(0001) surface disappears upon a Fe coverage of 2.5 ML, indicating that the growth proceeds via a 3D mechanism as previously reported by He *et al* [2]. The faint main streaks of Fe exhibiting a bcc structure start to emerge at a deposition of 8 ML. Clear RHEED streaks of the Fe film become unambiguous at the deposition of 10 ML from where onward, less intense reflexes are visible beside the main streaks and the Fe patterns appear spotty.

We further demonstrate that by reducing t_{Fe} down to a few ML, superparamagnetism (SPM) of the ultrathin Fe can be activated at the ambient temperature. Fig. 1(a) illustrates the hysteresis loop of an as-deposited 5 ML Fe(110) film on GaN(0001) taken at RT and reveals that the loop on one hand has an unsaturated magnetization and on the other hand possesses tiny but noticeable M_r and H_c. These two characteristics as a whole imply a coexistence of SPM and weak FM in the ultrathin film. In order to gain further insight into this mixed magnetic state, temperature dependence of the magnetization *M(T)* in the form of field cooling (FC) and zero field cooling (ZFC) curves of the 5 ML sample was measured by SQUID in a *T* range between 5 and 300 K as illustrated in Fig. 1(b). The ZFC magnetization curve was performed by applying a small in-plane field of 300 Oe to the sample at 5 K and then warming it with *M* being measured

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as a function of *T*. Similarly, the FC curve was done by cooling the sample to 5 K from 300 K in the presence of the same field strength. Notable in Fig. 6(b) is irreversibility of the ZFC and FC curves below 120 K, as indicated by the cusplike feature in the ZFC curve. This *T* value at which the *M* maximum occurs represents the blocking temperature T_B of the 5 ML Fe. More details will be given in the full manuscript.

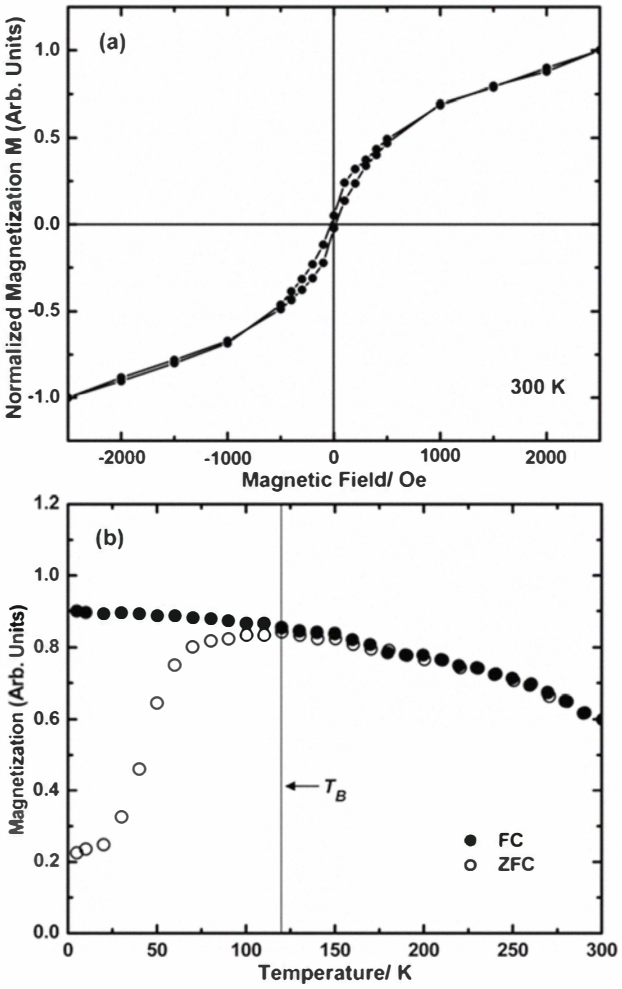

Fig.1 Magnetic hysteresis loop of a 5 ML thick Fe(110) film with an external field of 300 Oe applied along the [$1\bar{1}20$] direction of the GaN(0001) at RT and (b) the corresponding ZFC and FC magnetization curves. The vertical line in (b) refers to the estimated blocking temperature T_B of the Fe/GaN sample.

References

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